1,007,302

PATENT SPECIFICATION

DRAWINGS ATTACHED

1,007,302

Date of Application and filing Complete Specification June 12, 1964.
No. 24516/64.
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Index at acceptance: —C3 R(3D1, 3D13, 3D14, 3D20, 22N2, 22P3); B1 X3X6

Int. Cl.: —C 08 g

COMPLETE SPECIFICATION

Improvements in and relating to the Production of Thermoplastic Polycondensation Products

We, FARBENFABRIKEN BAYER AKTIENGESELLSCHAFT, a body corporate organised under the laws of Germany of, Leverkusen, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention is concerned with improvements in and relating to the production of thermoplastic polycondensation products.

The worm conveyor provides for a uniform transport of the reaction mixture through the tube and for a good mixing of the reaction material. By keeping free the inner space of the tube, which may be under reduced pressure, and by the continuous good mixing of the reaction material, which passes in a more or less thin layer along the inner wall of the tube from the inlet side to the exit side, there is also produced a comparatively large

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The inventors of this invention in the sense of being the actual devisers thereof within the meaning of Section 16 of the Patents Act, 1949 are HERMANN SCHNELL, Franz-Stollwerck Strasse 15, Krefeld-Uerdingen, Germany. RUDOLF SCHNEIDER, Bunsenstrasse 21, Krefeld, Germany; GOTTERIED GERLACH, Scheibler-strasse 83, Krefeld, Germany; and OTTO COURT, Deutsche Strasse 8, Neuss, Germany; all of German nationality.

THE PATENT OFFICE

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off from this free central part of the tube. If necessary, the pipes for leading off of the gases and/or vapours can be connected to a distillation column.

Depending on expediency, the reaction tube can be brought to different temperatures at different places. It is frequently advantageous to allow the temperature to increase from the inlet side to the exit side. The tube can be arranged vertically, horizontally or tilted.

In the new process according to the present invention, the heat necessary for the course of the polycondensation, as well as for the evaporation of the volatile reaction products split off and possibly of the solvent, can be transmitted easily and uniformly to the reaction mixture, without overheating occurring.

polycondensation, can possibly be highly viscous, with a residence time of only a few minutes, for example, of about 2—20 minutes.

The inner diameter and the length of the tube, the pitch of the worm conveyor, its speed of rotation, the loading of the apparatus, the course of the polycondensation reaction and the properties of the polycondensate must be harmony with one another. The most favourable relationships can be easily determined by single experiments. In general, good results are obtained with a pitch of the worm conveyor of about 10—200 mm. and with a speed of rotation of about 10—150 r.p.m., in the case of a tube of, for example, 100—200 mm. inner diameter and 400—2000 mm. length and a loading of

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The present invention is concerned with improvements in and relating to the production of thermoplastic polycondensation products which proceed with the separation of volatile reaction products and possibly of solvents.

The process according to the present invention consists in that viscous starting materials or pre-condensates or solutions thereof are conducted to the point of entry of a heated tube, which is possibly under reduced pressure, and passed along the inner wall of the tube by a conveyor worm (spiral), which may be heated, to the point of exit where the product is removed, the loading of the tube being so adjusted that the central part of the tube remains free and the volatile reaction products split off, and possibly solvent vapours, are led off from this free central part of the tube. If necessary, the pipes for leading off of the gases and/or vapours can be connected to a distillation column.

Depending on expediency, the reaction tube can be brought to different temperatures at different places. It is frequently advantageous to allow the temperature to increase from the inlet side to the exit side. The tube can be arranged vertically, horizontally or tilted.

In the new process according to the present invention, the heat necessary for the course of the polycondensation, as well as for the evaporation of the volatile reaction products split off and possibly of the solvent, can be transmitted easily and uniformly to the reaction mixture, without overheating occurring. The worm conveyor provides for a uniform transport of the reaction mixture through the tube and for a good mixing of the reaction material. By keeping free the inner space of the tube, which may be under reduced pressure, and by the continuous good mixing of the reaction material, which passes in a more or less thin layer along the inner wall of the tube from the inlet side to the exit side, there is also produced a comparatively large and constantly renewed phase boundary between the liquid reaction mixture and the gas or vapour space in the inner part of the tube, whereby the volatile reaction products split off are drawn off extremely quickly from the liquid reaction mixture.

These advantages render possible the production of polycondensation products of very high and uniform quality in a surprisingly short time. Thus, with the help of the new process, there can even be produced high molecular polycondensation products, for example, polyamides, polyesters and, amongst these, for example, polycarbonates and polyester-amides, with comparatively high molecular weights, the melts of which, in the end stage of the polycondensation, can possibly be highly viscous, with a residence time of only a few minutes, for example, of about 2—20 minutes.

The inner diameter and the length of the tube, the pitch of the worm conveyor, its speed of rotation, the loading of the apparatus, the course of the polycondensation reaction and the properties of the polycondensate must be harmony with one another. The most favourable relationships can be easily determined by single experiments. In general, good results are obtained with a pitch of the worm conveyor of about 10—200 mm. and with a speed of rotation of about 10—150 r.p.m., in the case of a tube of, for example, 100—200 mm. inner diameter and 400—2000 mm. length and a loading of

[*Price 4s* [redacted]

1.0—30 kg./hour. Of course, the device is capable of enlargement in any desired manner and is to be adapted to the desired rate of passage.

In some cases, it can be advantageous to introduce low molecular weight precondensates into the reaction tube, instead of the monomeric starting materials, i.e. for example ω-amino- or ω-hydroxy-carboxylic acids, or mixtures of diamines or dihydroxy compounds and dicarboxylic acids or dicarboxylic acid halides, or dicarboxylic acid diesters or diesters of carbonic acid, whereby the precondensates, the production of which is, in general, very much more easy to carry out than the end stage of the polycondensation, can be produced in any desired manner, for example, in a stirrer kettle or in a continuously-operating reaction tube.

The accompanying drawing shows the important features of the device to be used for the process, of the present invention, two examples being given. The reaction tube (1) is heated externally. The worm conveyor (2) present therein is set in rotation by the drive (3) and under takes the transport of the material supplied at the entry (4) to the exit (5), removal taking place by a geared pump (6) (Figure 1) or a worm (7) (Figure 2) but also by other known devices. The gases and/or vapours are led off *via* the pipes (8).

The following Examples are given for the purpose of illustrating the present invention:—

EXAMPLE 1:

Into a tube, heated externally to 270—300° C., of 100 mm. diameter and 600 mm. length, there is continuously passed, *via* a geared pump, 5.0 kg./hour of a precondensate heated to 250° C., which is produced from diphenyl carbonate and bisphenol A in the molar ratio 1.03 : 1.0 and has a relative viscosity of 1.100 measured in a 0.5% solution in methylene chloride at 20° C., and conveyed to the exit with a speed of rotation of the worm conveyor of 20 r.p.m. and under a pressure of 1 mm. Hg., with a mean residence time of 8—10 minutes, whereby the relative viscosity of the finished polycondensate increases to 1.350. 1.5% of split-off reaction products, referred to the amount of precondensate introduced, collect in the receiver.

EXAMPLE 2:

Into the same device as in Example 1, but at temperatures of 270—280° C. and a speed of rotation of the worm conveyor of 40 r.p.m., a material produced by the precondensation of terephthalic acid bis-(ethylene glycol) ester is passed at a rate of 1.8 kg./hour and further condensed at a mean residence time of 8 minutes, whereby the relative viscosity, measured in a 0.5% solution in a mixture of 40% tetrachloroethane and 60% phenol at 20° C., increase from 1.080 for the precondensate to 1.280 for the finished product.

WHAT WE CLAIM IS:—

1. Process for the production of thermoplastic polycondensation products, which proceeds with the separation of volatile reaction products and possibly of solvent, wherein viscous starting materials or precondensates or solutions thereof are conducted to the point of entry of a heated tube and moved by a worm conveyor along the inner wall of the tube to the point of exit where the product is withdrawn, the loading of the tube being so adjusted that the central part of the tube remains free and the volatile reaction products split off, and possibly solvent vapours, are led off from this free central part of the tube.

2. Process according to claim 1, wherein the heated tube is under reduced pressure.

3. Process according to claim 1 or 2, wherein the worm conveyor is heated.

4. Process according to claim 1 for the production of thermoplastic polycondensation products, substantially as hereinbefore described and with reference to Figure 1 or Figure 2 of the accompanying drawings.

5. Thermoplastic polycondensation products, whenever prepared by the process according to any of claims 1 to 4.

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1 SHEET *This drawing is a reproduction of the Original on a reduced scale*

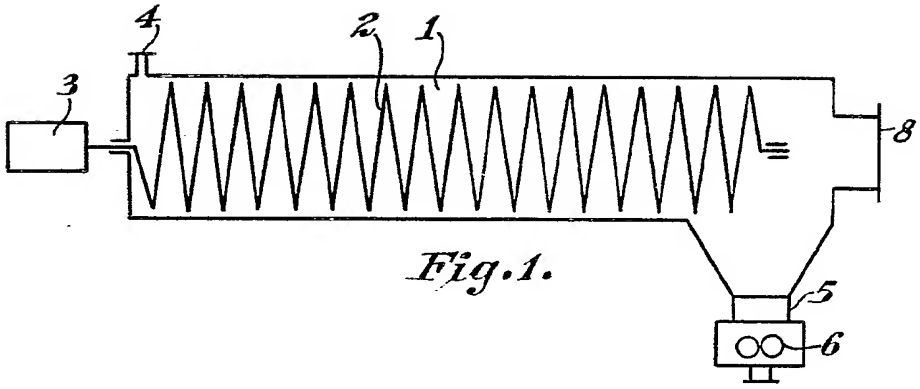

Fig. 1.

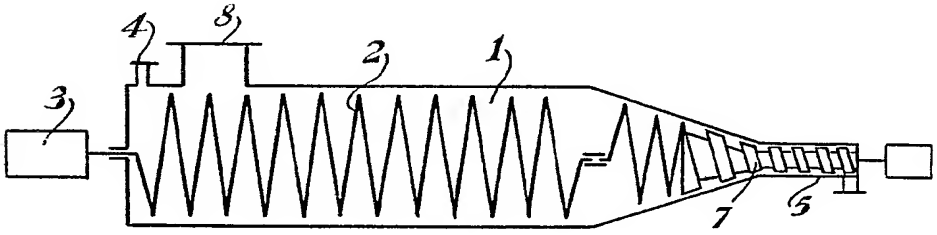

Fig. 2.